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Nonlinear Effects in the Dynamic Structure Factor of the Classical Antiferromagnetic Heisenberg Chain in an External Field at Low Temperatures

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Is is found that nonlinear effects give rise to anomalies in the spectrum of the antiferromagnetic Heisenberg chain in an external field, even at low temperatures. Divergences in the two-magnon density of states are responsible for additional resonances, besides the usual spin-wave peak, in the dynamic structure factor.

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Nonlinear effects in the dynamics of classical one-dimensional (1D) magnets, such as spin-energy coupling[1] or solitary solutions,[2] have received much attention in recent years. Apart from a broadening of the spin wave, the influence of the nonlinearity on the spectrum is not very clear. A second resonance in the spectrum of the longitudinal component of a Heisenberg chain, subject to a field, was found by Loveluck and Balcar[1] from a fit of a four-pole approximation to computer simulation data. Two of the present authors found second resonances for both components in the antiferromagnet, using a continued-fraction approach and starting from the exact knowledge of the six lowest frequency moments.[3] A complete understanding of the observed features could, however, not be obtained. Lovesey and Loveluck[4] argue that the second resonance is due to spin-energy coupling, whereas Reiter *et al.*[5] argue that a divergence in the two-magnon density of states causes this phenomenon. Although most theoretical studies are devoted to ferromagnetic chains, we think that the antiferromagnet deserves more attention, because it is of direct experimental relevance,[6-9] and because the competition between the exchange interaction and the magnetic field leads to very interesting static[10] and dynamic properties.[3] Therefore we examine this system from a different starting point, which promises to give more physical insight. The approach is due to Reiter and Sjölander, who applied it to the zero-field case, and it yields exact results in the low-temperature limit.[11]

Following Mori,[12] we can write for the Laplace-transformed spin-correlation function

$$C_q{}^\alpha(z) = C_q{}^\alpha(t=0)\frac{z+\Sigma_q{}^\alpha(z)}{z^2-\langle\omega^2\rangle_q{}^\alpha + z\Sigma_q{}^\alpha(z)},\quad z=\omega+i\epsilon,\quad \alpha=\perp,z, \tag{1}$$

where $\langle\omega^2\rangle_q{}^\alpha$ is the second frequency moment, which can be calculated rigorously with use of the transfer-operator method.[10] The difficult point is the calculation of the memory function $\Sigma_q{}^\alpha(z)$, which is defined as

$$\Sigma_q{}^\alpha(z) = -\langle\dot{S}_{-q}{}^\alpha\dot{S}_q{}^\alpha\rangle^{-1}\langle Q\ddot{S}_{-q}{}^\alpha[(z-QLQ)^{-1}Q\ddot{S}_q{}^\alpha]\rangle, \tag{2}$$

where $L=-i\,d/dt$ is the Liouville operator, and $Q=1-P$, with P projecting onto the spin density $S_q{}^\alpha$ and the spin-current density $\dot{S}_q{}^\alpha$. For $T\to 0$, $(\langle\omega^2\rangle_q{}^\alpha)^{1/2}$ tends to the spin-wave frequency $\omega_\alpha(q)$ and the memory function disappears in proportion with T. Reiter and Sjölander showed that to lowest nonvanishing order in T, the time evolution QLQ may be replaced by the ordinary time evolution L in Eq. (2) in the zero-field case.[11] Their arguments apply equally well when a field is present and we refer to their papers[11] for a detailed proof. The calculation of the memory function is then reduced to the calculation of

$$M_q{}^\alpha(z) = -\langle\dot{S}_{-q}{}^\alpha\dot{S}_q{}^\alpha\rangle^{-1}\langle Q\ddot{S}_{-q}{}^\alpha[(z-L)^{-1}Q\ddot{S}_q{}^\alpha]\rangle. \tag{3}$$

Because we only need the lowest order in T, $M_q{}^\alpha(z)$ can be calculated in the harmonic approximation. This calculation is very tedious, but straightforward in principle, and we merely give the essential steps.

Written in spherical coordinates, the Hamiltonian reads

$$H=\sum_n[\cos\theta_n\cos\theta_{n+1}-\sin\theta_n\sin\theta_{n+1}\cos(\xi_n-\xi_{n+1})-h\cos\theta_n],\tag{4}$$

with $S_n^x=(-1)^n\sin\theta_n\cos\xi_n$, $S_n^y=(-1)^n\sin\theta_n\sin\xi_n$, $S_n^z=\cos\theta_n$, and where the exchange constant $J=-1$. The ground state is obtained for $\cos\theta_n=\cos\theta\equiv\frac{1}{4}h$ and $\xi_n=\xi$, where ξ can be chosen arbitrarily. Defining ψ_n and φ_n as the deviations from equilibrium, expanding Eq. (4) in ψ_n and φ_n and then Fourier transforming, we obtain the Hamiltonian in the harmonic approximation,

$$H=E_0+\tfrac{1}{2}\sum_k[a(k)\psi_k\psi_{-k}+b(k)\varphi_k\varphi_{-k}].\tag{5}$$

The spin-wave dispersions can be expressed in $a(k)$ and $b(k)$ in the following way:

$$\omega_z(q)=[a(q)b(q)]^{1/2},\qquad \omega_\perp(q)=\omega_z(q^*),\quad q^*=\pi+q,\tag{6}$$

$$a(q)=2[1-\cos(2\theta)\cos q],\qquad b(q)=2(1-\cos q).\tag{7}$$

Note that the dispersion of the transverse component is the dispersion of the longitudinal component, shifted over half of the Brillouin zone. This is a consequence of the antiferromagnetic ordering of the transverse component in the ground state. In real space this is expressed by the factor $(-1)^n$ in the transformation given above. In order to obtain $M_q^\alpha(z)$, we have to express $Q\ddot{S}_q^\alpha$ in the harmonic coordinates ψ_k and φ_k, and we need to retain only the lowest-order terms, which are quadratic in ψ_k and φ_k. In the following we will limit ourselves to the transverse component because here the effect of the nonlinearity of the equations of motion is generally larger than for the longitudinal component.

We now give the result for the imaginary part of the memory function. The real part can then be obtained with use of the Kramers-Kronig relations. We have

$$\mathrm{Im}[\Sigma_q^\perp(\omega)]=\frac{2T\sin^2\theta(1+\cos q^*)}{\cos^2\theta(1-\cos q^*)+1-\cos(2\theta)\cos q^*}\int_0^\pi dk\{[(P-Q)^2+(R+S)^2][\delta(\omega-\Omega_+)+\delta(\omega+\Omega_+)]+[(P+Q)^2+(R-S)^2][\delta(\omega-\Omega_-)+\delta(\omega+\Omega_-)]\},\tag{8}$$

with

$$\Omega_\pm=|2\sin(\tfrac{1}{2}k+\tfrac{1}{4}q^*)[a(k+\tfrac{1}{2}q^*)]^{1/2}\pm2\sin(\tfrac{1}{2}k-\tfrac{1}{4}q^*)[a(k-\tfrac{1}{2}q^*)]^{1/2}|,\tag{9}$$

$$P=1-(\cos\tfrac{1}{2}q^*)(\cos\tfrac{1}{2}q^*+\cos k),\tag{10}$$

$$Q=2\left[\cos\frac{q^*}{2}\sin^2\frac{q^*}{2}+\left(1+2\cos^2\theta\sin^2\frac{q^*}{2}\right)\cos k-\cos(2\theta)\cos\frac{q^*}{2}\cos^2k\right]\left[a\left(k+\frac{q^*}{2}\right)a\left(k\frac{q^*}{2}\right)\right]^{-1/2},\tag{11}$$

$$R=2\cos\theta[\sin(\tfrac{1}{2}k-\tfrac{1}{4}q^*)\cos k-\sin\tfrac{1}{2}q^*\cos(\tfrac{1}{2}k-\tfrac{1}{4}q^*)][a(k+\tfrac{1}{2}q^*)]^{-1/2},\tag{12}$$

$$S(k,q^*)=R(k,-q^*).\tag{13}$$

This result is exact to the extent that it contains all the frequency moments exact up to lowest nonvanishing order in the temperature. By a change of the integration variable in Eq. (8), it is easily seen that the memory function diverges whenever the two-magnon density of states $n_\pm=dk/d\Omega_\pm$ diverges. For $h=0$, we have $\theta=\pi/2$ and we reobtain the result of Ref. 11. We remark that for a given wave vector q, the memory function diverges for $\pm\omega=\Omega=4|\cos\frac{1}{2}q|$ and we see that Ω is always larger than the spin-wave frequency $\omega(q)=2|\sin q|$. The effect of these divergencies on the dynamic structure factor is generally not very important for $h=0$.[11] Analytic results can also be obtained for $h=2\sqrt{2}$, or equivalently $\theta=\pi/4$. In this case we find divergencies for $\Omega_1=2h\times|\sin\frac{1}{4}q^*|$ and $\Omega_2=2h|\cos\frac{1}{4}q^*|$. As for $h=0$, Ω_1 and Ω_2 are always larger than the spin-wave frequency, which is now given by $\omega_\perp(q)=h|\sin\frac{1}{2}q^*|$. A careful examination of Eq. (9) will show that by varying the magnetic field, very different situations can occur. In some cases there are two frequencies where the memory function diverges; in other cases, three or four. Furthermore, for magnetic fields that are not too large, divergencies occur at frequencies lower than the spin-wave frequency. In order to study these cases we have to calculate both the real and imaginary part of the memory function numerically. For

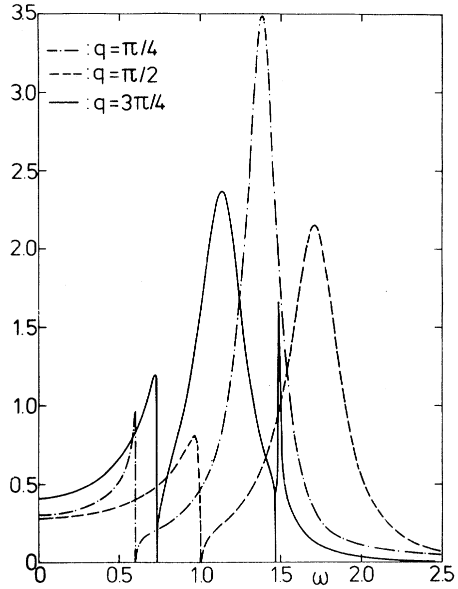

FIG. 1. The normalized dynamic structure factor for $h=1$ and $T=0.2$ and for different wave vectors. The frequency scale is the same as that of Fig. 2, but we have not plotted the whole range, because for higher frequencies the effects are too small to be seen on this scale.

this purpose we used the linear analytic method as proposed by Gilat.[13] The special case $h=2\sqrt{2}$, for which we have an analytic expression, provided a thorough check on the numerical results.

In Fig. 1 we have plotted the dynamic structure factor, which is the imaginary part of $C_q^{\perp}(z)$ as defined by Eq. (1), for $h=1$ and $T=0.2$ and for different wave vectors. In Fig. 2 we have plotted the imaginary part of the memory function for the same parameters. The spiky structure of the memory function is clearly reflected in the dynamic structure factor and it gives rise to additional peaks, besides the usual spin-wave resonance. Mostly, the weight of these resonances is rather small. However, when the memory function diverges at a frequency smaller than the spin-wave frequency, peaks bearing considerable weight may originate. It can be seen from Eqs. (1) and (8) that, as the temperature is lowered, the anomalies in the spectrum do not disappear, although the spin-wave resonance gains more weight. If we compare our results with the continued-fraction approach,[3] which is not restricted to low temperatures, we see that the agreement is only qualitative. The sharp structure, induced by the singularities, is smeared out in this approach. Nevertheless, a second resonance at the

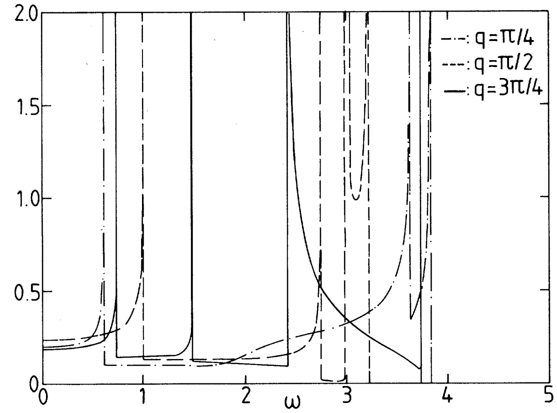

FIG. 2. The normalized imaginary part of the memory function for the same parameters as in Fig. 1.

left of the spin-wave peak is also observed in this approach, but most of the details of the spectrum are lost.

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